KOKAI PATENT APPLICATION NO. HEI 4-67577

TOTALLY SOLID ELECTROCHEMICAL ELEMENT

[Translated from Japanese]

[Translation No. LPX20157]

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JAPANESE PATENT OFFICE (JP)

PATENT JOURNAL (A)

KOKAI PATENT APPLICATION NO. HEI 4-67577

Technical Indication Section

Int. Cl.^{5}:	H 01 M 10/38
Identification code:	
Sequence Nos. for Office Use:	8939-4K
Filing No.:	Hei 2-180006
Filing Date:	July 6, 1990
Publication Date:	March 3, 1992
No. of Claims:	3 (Total of 4 pages in the Document)
Examination Request:	Not filed

TOTALLY SOLID ELECTROCHEMICAL ELEMENT

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[*There are no amendments to this patent.*]

Specification

1. Title of the invention

Totally solid electrochemical element

2. Claims of the invention

(1) A totally solid electrochemical element having at least a solid electrolyte layer and at least a pair of electrode layers arranged via the above-mentioned solid electrolyte layer wherein the size of the area of the above-mentioned solid electrolyte layer and the size of the area of the above-mentioned electrode layer are different.

(2) The totally solid electrochemical element described in Claim 1 wherein the solid electrolyte layer has the composition $4AgI \cdot Ag_2WO_4$.

(3) The totally solid electrochemical element described in Claim 1 wherein the electrode activation material in the electrode has the composition $Ag_xV_2O_{6-y}$ ($0.6 \leq x \leq 0.8$, y is the oxygen deficiency).

3. Detailed description of the invention

Field of industrial application

The present invention pertains to a totally solid electrochemical element that utilizes a solid electrolyte, such as a totally solid potential memory element and a solid electrolyte battery.

Prior art

Basically, a totally solid electrochemical element is a structure comprising three layers consisting of electrode/solid electrolyte/electrode, and in the past, totally solid electrochemical

elements were produced by pressure lamination of the above-mentioned three layers using a single molding die. For example, in production of a potential memory element, pressure molding is performed for a solid electrolyte in a molding die, electrode material is arranged at the top and bottom of the molding die, and solid molding is carried out under pressure and the element is removed from the die. In this manner, a totally solid electrochemical element comprising an electrode layer/solid electrolyte layer/electrode layer can be produced. Finally, the end member of the above-mentioned molding is chipped and lead terminals are bonded. Furthermore, sealing is performed with a resin, as needed.

Problems to be solved by the invention

In the production of the above-mentioned totally solid electrochemical element, the end member of the solid electrolyte layer is contaminated with the electrode material, etc. adsorbed after pressure molding, and short-circuiting of the two electrodes often occurs. Thus, chipping of the end member of the molding is required, which process requires time, and furthermore, possible destruction of the molding poses a problem at the time of chipping the end member.

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Means to solve the problem

In order to eliminate the above-mentioned conventional problem, the purpose of the present invention is to produce a totally solid electrochemical element where the area of the solid electrolyte layer is increased so that it is greater than the area of the electrode layers.

Work of the invention

The present invention has the above-mentioned structure, thus, adsorption of the electrode material powder on the inner walls of the die is absent at the time of molding, short-

circuiting between the electrodes arranged at the top and bottom of the solid electrolyte layer can be prevented, and furthermore, the chipping process for the end member of the molding can be eliminated.

Application example

A totally solid potential memory element used as an application example of a totally solid electrochemical element is explained in specific terms below.

<Application Example 1>

Amounts of AgI, Ag_2O and WO_3 were measured to form a molar ratio of 4:1:1, and mixing was done in an alumina mortar. Subsequently, a pressure molding of the above mixture was carried out, and it was injected into a Pyrex tube under reduced pressure, and the reaction was carried out at a temperature of 400°C for 17 hours. Subsequently, wet crushing was performed for the above-mentioned reaction product in a ball mill, classification was done to produce a silver-ion-containing, conductive solid electrolyte powder of $4AgI \cdot Ag_2WO_4$ of 200 mesh or below.

Subsequently, quantities of vanadium oxide V_2O_6 and metal silver powder were measured to provide a molar ratio of 1:0.7 and mixing was done in a mortar. Pressure molding was carried out for the above mixture, and it was injected into a quartz tube under reduced pressure and the reaction was carried out at a temperature of 600°C for 48 hours. The above reaction product was crushed in a mortar and classified to produce an electrode activation powder of vanadium silver oxide represented by the formula $Ag \cdot V_2O_6$.

With the solid electrolyte and electrode activation material produced above, production of potential memory element was carried out according to the method shown below. First, the solid

electrolyte powder and electrode activation material were mixed and production of an electrode material containing 30% of the electrode activation material in the electrode was carried out. A 20 mg quantity of the above electrode material was measured and pressure molding was carried out under a pressure of 2 ton/cm^2 to produce a molded electrode having a diameter of 5 mm. Subsequently, a 300 mg quantity of the solid electrolyte was measured and pressure molding was carried out under a pressure of 2 ton/cm^2 to produce an molded electrolyte with a diameter of 7 mm. The electrode molding and electrolyte molding pair produced as described above were arranged in the order of electrode molding/electrolyte molding/electrode molding and lamination of said materials was done under a pressure of 4 ton/cm^2 to produce the potential memory element shown in Fig. 1. In order to confirm the voltage retention characteristics of the above-mentioned potential memory element, copper wires plated with tin were bonded to the electrode layers on each side of the molding with a conductive carbon paste, and a powder coating was further applied to the entire molding with an epoxy resin at a temperature of 150°C.

Furthermore, production of a totally solid electrochemical element with a conventional structure was carried out according to the method explained below. First, the solid electrolyte powder and electrode activation material powder produced above were mixed to produce an electrode material containing 30% of the electrode activation material. First, a 300 mg quantity of the solid electrolyte was measured and pressure molding was carried out under a pressure of 2 ton/cm^2. Subsequently, a 30 mg quantity of electrode material was measured and inserted at the top and bottom of the die where molding of the solid electrolyte layer was being performed, and pressure molding was carried out under a pressure of 4 ton/cm^2. The above-mentioned molding was removed from the die, then, copper wires plated with tin were bonded to the electrode layers

on each side of the molding with a conductive carbon paste, and a powder coating was further applied to the entire molding with an epoxy resin at a temperature of 150°C.

100 each of the above-mentioned two different types of potential memory elements were produced and in order to check for short-circuiting at the terminal of the above-mentioned potential memory elements, constant voltage charging was carried out for 20 hours at 100 mV, and the terminal voltage of the potential memory elements was examined after 2 hours under an open circuit condition. Fig. 3 shows a comparison of the voltage stability of the conventional potential memory element and the potential memory element of Application Example 1 of the present invention. As shown in Fig. 3, in the case of the potential memory element having the conventional structure, a sharp decrease in the terminal voltage due to short-circuiting at the end of the molding is observed. On the other hand, reduction in the terminal voltage due to short-circuiting at the end of the molding is not observed in the potential memory element of Application Example 1.

Application Example 2

The solid electrolyte and electrode material used in Application Example 1 were used in this case as well, and production of a potential memory element was done according to the method described below. A 20 mg quantity of electrode material was measured and pressure molding was carried out under a pressure of 2 ton/cm^2 to produce an electrode molding with a diameter of 5 mm. Subsequently, a 300 mg quantity of solid electrolyte was measured and pressure molding was carried out under a pressure of 2 ton/cm^2 to produce the electrolyte molding shown in Fig. 2.

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Subsequently, the electrode moldings were placed in concave recesses formed at the top and bottom of the electrolyte molding and pressure molding was carried out under 4 ton/cm^2 to produce a totally solid electrochemical element having a diameter of 7 mm. In order to confirm the voltage retention characteristics of the above-mentioned potential memory element, a copper wire plated with tin was bonded with the electrode layers on each side of the molding with a conductive carbon paste, and a powder coating was applied to the entire molding with an epoxy resin at a temperature of 150°C.

A total of 100 of the potential memory elements of the present invention were produced as described above and the test described in Application Example 1 above was performed and the results obtained are shown in Fig. 4. As Fig. 4 shows, short-circuiting can be effectively prevented in the potential memory element having a cross-section shaped-like the letter H [sic] as in the case of the potential memory element of Application Example 1.

In this case, the shape of the solid electrolyte and electrode activation material is not limited to those shown in Fig. 1 or Fig. 2, and in addition to a disk-shaped molding, all types of polygonal moldings such as triangular and rectangular moldings can be used as long as the electrode molding material does not come in contact with the inner wall of the die for solid molding. Furthermore, in the above-mentioned application examples, a totally solid electrochemical element is used as an example, but the present invention can be effectively used for a totally solid electrochemical element such as total solid battery having a structure where a pair of electrodes are used with a solid electrolyte.

Effect of the invention

As described above, short-circuiting at the end member of the molded element can be

eliminated when the area of the solid electrolyte layer is increased with respect to the area of the electrode layer in a totally solid electrochemical element having a solid electrolyte layer and electrode layers on both sides, and chipping of the end member after molding can be eliminated, and damage to the molding based on the chipping process can be eliminated.

4. Brief description of figures

Fig. 1 is a perspective view that shows a potential memory element used as an application example of a the totally solid electrochemical element of the present invention; Fig. 2 is the perspective view that shows a different example of a potential memory element of the present invention; Fig. 3 shows the voltage retention characteristic of the potential memory element of Application Example 1, and Fig. 4 shows the voltage retention characteristic of the potential memory element of application example 2.

1 .. Electrode layer, 2 .. Solid electrolyte layer, 3 .. Electrode layer, 4 .. Electrode layer, 5 .. Solid electrolyte layer, 7 .. electrode layer.

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Fig. 1

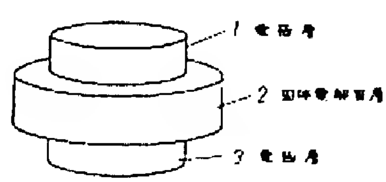

Fig. 2

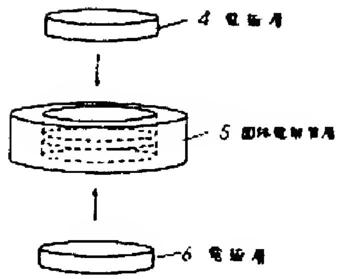

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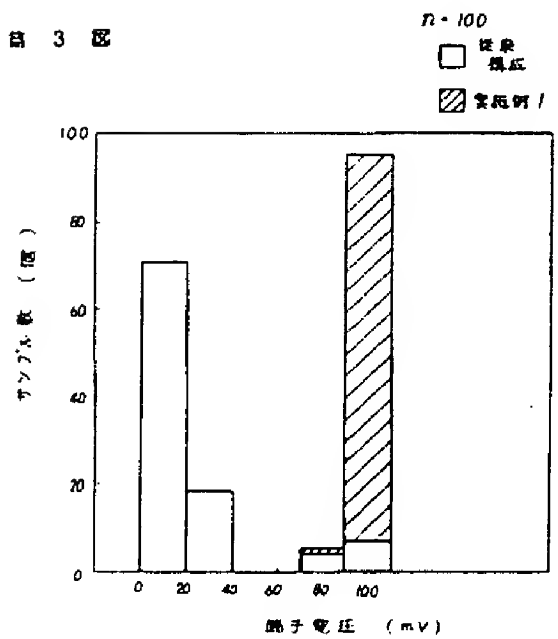
Fig. 3



Solid: Conventional structure Shaded: Application example 1

Vertical axis: Number of samples (pieces) Horizontal axis: Terminal voltage (mV)

Fig. 4

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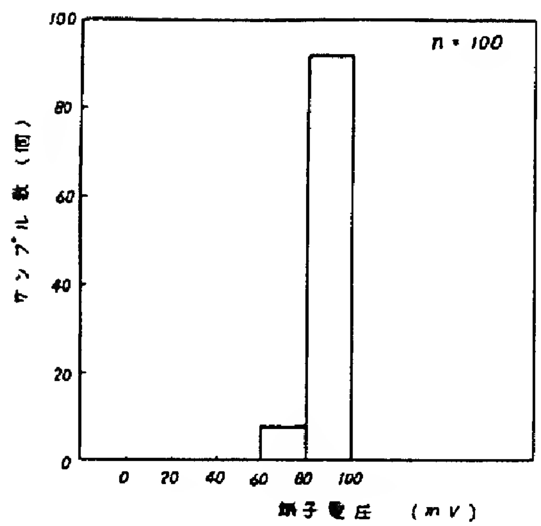

Vertical axis: Number of samples (number)

Horizontal axis: Terminal voltage (mV)